

Charge Density Wave in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$

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Recent experimental results [1,2] revealed the stability of a peculiar charge density wave (CDW) with odd period ($\lambda = 3, 5$) in the coupled Cu_2O_5 spin ladders in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$, while the CDW with even period is unstable. Here we resolve this contradiction by solving the effective t - J model for the two coupled chains supplemented by the intersite interaction term V .

Crucial to this finding is the novel interaction term V (responsible for the interladder repulsion between nearest neighbor holes), which is derived from the on-site Hubbard-like repulsion between electrons on the same oxygen sites but within two distinct orbitals which belong to two neighboring Cu_2O_5 ladders in the charge transfer model [3].

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1 Introduction It is at the heart of the understanding of the high-temperature superconducting (SC) phase to infer what kind of state competes with the SC one in the possible quantum phase transition between the two. As in the two-dimensional (2D) copper oxides one of the candidates of such a state is the stripe phase, it is natural to expect that in the quasi-2D copper oxide coupled-ladder system also a kind of charge ordered state would compete with the predicted there SC state [4]. Indeed in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$, which has the SC state for $x = 13.6$ and under pressure larger than 3 GPa [5], a charge density wave (CDW) phase with period $\lambda = 3$ and $\lambda = 5$ was observed when the Calcium concentration was tuned to $x = 11$ (which corresponds to the hole concentration $n_h = 6/5$ per copper) and $x = 0$ (where $n_h = 4/3$), respectively [1,2]. Although the charge transfer model for coupled ladders proposed in Ref. [3] may explain the onset of such an odd-period CDW state, it is the t - J -like model which is a *natural* Hamiltonian for the doped *spin* ladders [1,2]. However, then this experimental observation is a challenge for the theory since such a novel CDW state with the odd period has not been predicted by the standard t - J model for the ladders [6]. Moreover, the standard t - J model predicts the CDW order with period $\lambda = 4$ to be stable for $n_h = 5/4$ which would correspond to the $x = 4$ doping in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ where no charge order was observed [1,2].

The purpose of this paper is to shed some light on this apparent contradiction between the experiment and the theory. Therefore, first we discuss which terms are missing in the standard t - J model while in the second part of the paper we look at the generic features of this new model by solving it in a simplified case for two coupled chains using the Gutzwiller approach and the mean-field approximation.

2 Model Our starting point is the effective one-dimensional (1D) t - J model for a single chain,

$$H = -t \sum_{i\sigma} \left(\tilde{d}_{i\sigma}^\dagger \tilde{d}_{i+1,\sigma} + \text{H.c.} \right) + J \sum_i \left(\mathbf{S}_i \cdot \mathbf{S}_{i+1} - \frac{1}{4} \tilde{n}_{id} \tilde{n}_{i+1,d} \right), \quad (1)$$

where: (i) the operator $\tilde{d}_{i\sigma}^\dagger = d_{i\sigma}^\dagger (1 - n_{id,-\sigma})$ creates an electron at site i with spin σ in the restricted Hilbert space with no double occupancies, (ii) $\tilde{n}_{id\sigma} = \tilde{d}_{i\sigma}^\dagger \tilde{d}_{i\sigma}$ is the corresponding electron number operator, and (iii) \mathbf{S}_i is an $S = 1/2$ spin operator. Besides, the average number of d holes per site here is $n = 2 - n_h$, with $n = \sum_\sigma \langle \tilde{n}_{id\sigma} \rangle$.

In the standard approach to the Cu_2O_5 systems this model is derived from the charge transfer model which is similar to that for CuO_2 planes [7]. Then the model should also contain the kinetic $\propto t$ and superexchange $\propto J$ terms

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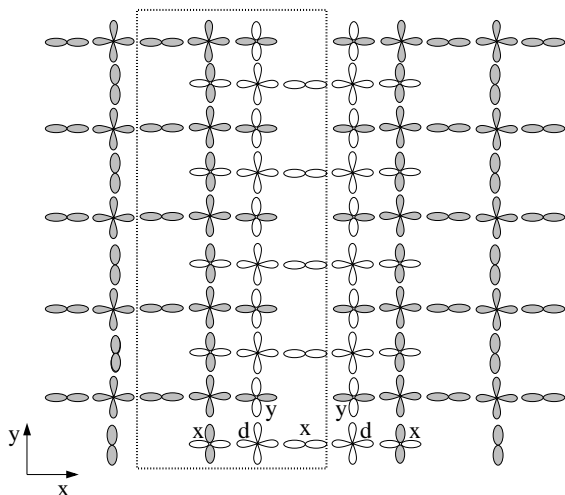


Figure 1 Artist’s view of the three coupled Cu_2O_5 ladders (denoted by grey, white, and again grey color, respectively). Only orbitals which are included in the charge transfer model of Ref. [3] are shown (where $x \equiv 2p_x$, $y \equiv 2p_y$, and $d \equiv 3d_{x^2-y^2}$). The vertical box depicts two coupled CuO_3 chains belonging to two different neighboring ladders and discussed in the paper.

along the rungs of the ladder — it would then constitute the standard t - J model for the ladder [6]. However, a closer look at the geometry of the Cu_2O_5 coupled ladders reveals that even such a model would not be enough. In fact, one cannot neglect a Coulomb repulsion on-site element between two holes on the same oxygen sites ($\propto U_p \gg t_{pd}$ [8], where t_{pd} is the Cu–O hybridization) but on two different oxygen $2p$ orbitals (x and y orbitals in Fig. 1) belonging to the two neighboring chains in different ladders [3]. Thus, although the holes do not hop between the neighboring Cu_2O_5 ladders (a small inter-oxygen hopping can be neglected [8]) the t - J model for the single ladder should contain an extra *interladder* coupling which can be derived from this Coulomb interaction.

Altogether this means that neglecting the kinetic and superexchange terms along the rung connecting the two chains and taking into account the interaction between the two neighboring chains is *a priori* as justified in the treatment of Cu_2O_5 coupled ladders as skipping the interladder coupling and considering merely the t - J model for a single ladder. Naturally, the proper approach is to take into account both of these effects (under which the work is in progress) but in what follows we merely concentrate on the problem of two coupled chains connected by this interladder coupling, see Fig. 1. This will enable us to study the generic role of the interladder coupling.

While the detailed derivation of the interladder coupling is somewhat lengthy and will be described elsewhere, a simple cartoon-picture shown in the left panel of Fig. 2 explains its basic idea. Here we see that the two Zhang-

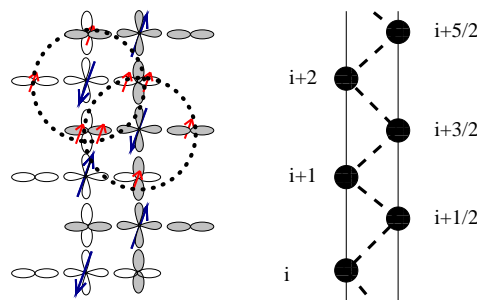


Figure 2 Left panel: the artist’s view of the interladder repulsion between two nearest neighbor Zhang-Rice singlets on two nearest neighbor chains. The Zhang-Rice state is depicted by a dotted ring. Large (small) arrows depict the hole spins for +1.0 (+0.25) charge. Right panel: the geometry of the model described by the $\tilde{H} + \tilde{H} + H'$ Hamiltonian, see text for further details.

Rice singlets [9] which are situated next to each other in the two neighboring chains (see dotted rings in Fig. 2) share the same oxygen sites. More precisely, as an oxygen hole forming a Zhang-Rice singlet state is equally distributed among the four oxygen sites surrounding the central copper hole [9], there is a 0.25 hole charge in each of these oxygen orbital sharing the common oxygen sites. Since the two holes in two different orbitals repel as $\propto U_p$ (where the Hund's exchange J_H is skipped for simplicity), the Zhang-Rice singlets repel as $\propto (1/4 \times 1/4 + 1/4 \times 1/4)U_p$. Obviously, the Zhang-Rice states in each chain are not the proper orthogonal states (similarly as in the 2D case [9]) and one needs to orthogonalize them first. This, however, does not change the result qualitatively as it occurs that such a procedure yields that the two Zhang-Rice singlets repel as $\propto 0.14U_p$. Besides, finite Hund's exchange reduces this interaction by a factor $(1 - 5J_H/2U_p)$.

Hence, we supplement the t - J model for a single chain, Eq. (1), by the following interladder term which couples the two chains under consideration

$$H' = V \sum_i \left(\tilde{n}_{id} \tilde{n}_{i+\frac{1}{2},d} + \tilde{n}_{id} \tilde{n}_{i-\frac{1}{2},d} \right), \quad (2)$$

where $V = 0.14U_p(1 - 5J_H/2U_p)$ and for realistic parameters [3] $V \sim 0.5t$. Here the *bar* signs are added over the operators which act in the Hilbert subspace of the neighboring chain. The detailed geometry of the full model for two coupled chains $\mathcal{H} \equiv H + \bar{H} + H'$ can be seen in the right panel of Fig. 2 — the solid lines show the bonds along which the kinetic and superexchange term of Eq. (1) act, while the dashed lines show the bonds along which the interladder terms of Eq. (2) are finite.

3 Results In order to solve the t - J - V model, given by the Hamiltonian \mathcal{H} , we first need to overcome the constraint of no double occupancies. One of the approximate

ways to do it is to introduce the Gutzwiller factors $g_t = (2 - 2n)/(2 - n)$ and $g_J = 4/(2 - n)^2$, which renormalize the kinetic term (g_t) and the superexchange and interladder terms (g_J). Similar factors were used recently to investigate the superconducting flux phases in the cuprates [10]. Then the whole Hamiltonian is defined in terms of unrestricted fermions $d_{i\sigma}^\dagger$.

Next, we decouple the fermion operators in the superexchange and interladder terms in a mean-field way: $d_{i\sigma}^\dagger d_{i\sigma} d_{i\sigma'}^\dagger d_{i\sigma'} \rightarrow d_{i\sigma}^\dagger d_{i\sigma} \langle d_{i\sigma'}^\dagger d_{i\sigma'} \rangle + \langle d_{i\sigma}^\dagger d_{i\sigma} \rangle d_{i\sigma'}^\dagger d_{i\sigma'}$ (a similar decoupling is used also for the \bar{b} operators). Then, we diagonalize the effective one-particle Hamiltonian in k space assuming that the classical fields take their initial values as

$$\langle d_{i\sigma}^\dagger d_{i\sigma} \rangle = \begin{cases} n - p & \text{for } i/\lambda \in \mathbb{Z} \\ n + \frac{1}{\lambda-1}p & \text{for } i/\lambda \notin \mathbb{Z} \end{cases}, \quad (3)$$

which defines the CDW order parameter p . As already mentioned, here λ is the CDW period, cf. Fig. 1 in Ref. [3]. In addition, $\langle \bar{d}_{i-\frac{1}{2},\sigma}^\dagger \bar{d}_{i-\frac{1}{2},\sigma} \rangle$ are defined as in Eq. (3) but with i/λ replaced by $(i+1)/\lambda$ [($i+2$)/ λ for period $\lambda = 5$] – such assumption minimizes the classical energy cost of the interladder repulsion V . Finally, the actual values of the classical fields (and consequently the order parameter p) are obtained by iterating the above procedure (i.e. calculated from the diagonalized Hamiltonian) until their initial and final values converge.

As the main result of this self-consistent procedure, we obtained the increasing order parameter p as a function of the interladder interaction V for a realistic value of $J = 0.4t$ [8] and for the three interesting hole dopings, see Fig. 3. First, we see that for finite values of V the CDW phase with the peculiar odd period is stable. Moreover, it is stable for realistic values of parameter $V \sim 0.5t$. Second, the CDW order with period $\lambda = 4$ is less stable as the CDW order sets in only for $V > 0.6t$.

One may ask, why the CDW phase with even period appears to be less stable. Although this reproduces the experimental result quite well, it is a very peculiar phenomenon associated purely with the 1D physics. In fact, it is the CDW phase with period $\lambda = 3$ which is remarkably stable here due to the nesting of the band structure for this particular doping, and indeed we find the exponential growth of p with increasing $V > 0$, special for the present 1D model.

4 Conclusions In summary, we showed that including the Coulomb repulsion between holes on the same oxygen sites but in different orbitals and belonging to two different neighboring Cu_2O_5 ladders, led to the effective repulsion between Zhang-Rice singlets which in turn led to the interladder repulsion V between nearest neighbor holes situated in the neighboring ladders. Next, we solved a t - J model supplemented by the repulsive term V in a simplified geometry of two coupled chains. It occurred that the CDW state with odd period was stable in such a model for

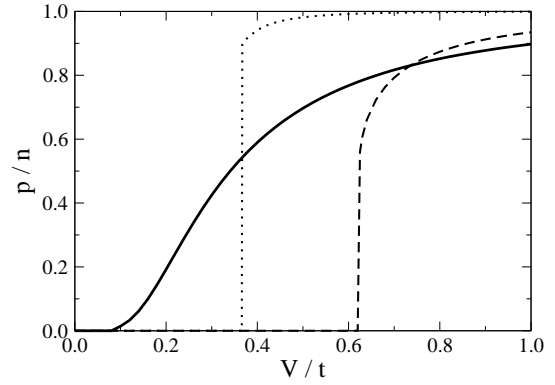


Figure 3 The CDW order parameter p as a function of the interladder interaction V in the two coupled chains with $J = 0.4t$: for filling $n = 2/3$ ($n_h = 4/3$) with period $\lambda = 3$ (solid line), for filling $n = 3/4$ ($n_h = 5/4$) with period $\lambda = 4$ (dashed line), and for filling $n = 4/5$ ($n_h = 6/5$) with period $\lambda = 5$ (dotted line).

the realistic values of the interaction parameters V , while the CDW state with the even period ($\lambda = 4$) was stable merely for slightly enhanced values of these parameters.

The results presented in this paper could possibly explain the onset of the novel odd-period-CDW state in the Cu_2O_5 coupled ladder system in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ with $x = 0$ and $x = 11$. However, a detailed study of this phenomenon is currently under investigation since one needs to generalize the t - J - V model presented here to the coupled ladders and to investigate the stability of CDW states in this generalized model in order to verify this conjecture.

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